The magnetoelastic excitation spectrum in the rare earth pyrochlore Tb₂Ti₂O₇

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I. SAMPLE COMPARISON

In this study we have used two different samples of Tb₂Ti₂O₇, and here we compare quantities such as their lattice parameter, specific heat and inelastic neutron scattering response. We have suggested elsewhere¹, and in the article that the inelastic neutron scattering responses that we study are not sample dependent properties, and here we show that to be the case.

A. Lattice Parameter

As in Yb₂Ti₂O₇², a useful diagnostic feature of a Tb₂⁺ₓTi₂₋ₓO₇₋ₙ sample should be the lattice parameter a, which can be compared to the trends of a vs x published in Refs. 3–5, though the practise of comparing Tb₂Ti₂O₇ samples by lattice parameter has recently been criticised and it has been suggested that different batches of Tb₂Ti₂O₇ (which is a material of poorly defined stoichiometry) used in different laboratories underlie the sample dependent properties. The lattice parameters of samples A and B were reported in the previous works⁶,⁷. They are a = 10.155288(1) Å and a = 10.1528(5) Å respectively. If the lattice parameter is taken as the main indicator of the stoichiometry parameter x in Tb₂⁺ₓTi₂₋ₓO₇₋ₙ by comparison to the trend published in Ref. 3, we see that the two samples fall on opposite sides of the compositional range in which a sharp peak in the heat capacity was experimentally observed, with xₐ ≈ 0.013 and xₜ ≈ −0.007⁷.

Many of the sample-dependent reports in the literature are studies of single crystals, and although crystals of controlled composition now appear to be possible⁸, it has been pointed out that large crystals may be inhomogeneous. Indeed, in one case, several points of the composition phase diagram were realized along the length of a single crystal boule⁹. However, this crystal has an obvious compositional gradient (as manifested in a changing

colour from one end to the other), and it has been pointed out elsewhere that the distribution of lattice strain in a typical large Tb₂Ti₂O₇ crystal (actually sample A) is not larger than in other rare earth pyrochlores that are not thought to suffer from such effects¹⁰.

B. Specific Heat

The specific heat of sample A was originally reported in Refs. [1,6]. The sharp peak at T = 0.47 K is not present in this sample. The heat capacity of sample B was measured by a procedure identical to that described in Refs. [1,6]. We see that sample B does have a sharp heat capacity peak. The heat capacities are otherwise identical, as shown in Fig. 1.
C. Comparison of diffuse and inelastic neutron scattering

Diffuse scattering data for both samples has been published in Refs. 11 and 12. The experiments are not simply comparable since the former employs polarization analysis and integrates all fluctuations within its energy window, while the latter is unpolarized but discriminates the elastic scattering from quasielastic and inelastic contributions by using a rather sharp energy resolution (0.07 meV). However, polarized inelastic measurements in Ref. 11 suggest that the diffuse scattering observed there is completely dominated by elastic scattering, and the sum of the polarized channels appears to resemble quite closely the unpolarized diffuse scattering reported in Ref. 12. At this level of comparison, the diffuse scattering reported for these samples is also highly similar to other samples reported in the literature by other groups. Recently, using several crystals of known values of $x$ and associated heat capacity behaviors, it has been shown that the diffuse scattering is not a sample dependent property.

Both samples have previously been measured on the time-of-flight spectrometer IN5 at a wavelength of 4 Å, which gives considerable detail of the dispersion and structure factor of the first crystal field level. We do not have identical coverage of $S(\vec{Q}, \omega)$, and different sample environments were employed, but where the two experiments can be compared, we find that the lineshape and dispersion of the exciton branches which can be distinguished in an unpolarized measurement are identical. There are small differences in intensity as a function of $\vec{Q}$ and $\hbar\omega$, but these are most plausibly due to the different sample geometries or sample environments. In Fig. 2 we show some salient comparisons.

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FIG. 2: Comparison of cuts through $S(\vec{Q}, \omega)$ of the two samples, measured on IN5

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